18p-D7-3

Photocarrier recombination dynamics in Cu_2ZnSnS_4 single crystals

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The optimal band-gap energy for solar energy conversion and the high absorption coefficients of the earth-abundant quaternary compound Cu_2ZnSnS_4 (CZTS) make it an ideal material for future solar cell applications [1-3]. However, the power conversion efficiency of CZTS-based solar cells is reported to be relatively low [4], which is thought to be directly related to intrinsic defects in CZTS [5-7]. Thus, to improve the efficiency, it is vital that the optical response of and the photocarrier recombination mechanisms in CZTS are well understood [8].

In this report, we clarify the temperature-dependent photoluminescence (PL) dynamical properties in CZTS single crystals based on a combination of experimental results obtained from simultaneous steady-state and time-resolved PL measurements. We found a significant change of nearly four orders of magnitude of the PL decay time, from microseconds at low temperatures to subnanoseconds at room temperature. The slow PL decay at low temperatures indicates localization of the photocarriers at the band tails. Drastically enhanced nonradiative recombination plays a crucial role in the photocarrier recombination dynamics at high temperatures, and consequently leading to the significant decrease of the PL decay time as well as the considerable quenching of the PL intensity. We show that the fast nonradiative recombination process is one of the main factors that ultimately determine the power conversion efficiency of CTZS-based solar cells. Our results, therefore, provide essential knowledge for further improving the performance of CZTS-based solar cells.

Part of this work was supported by JST-CREST and the Sumitomo Electric Industries Group CSR Foundation.

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